Test Results of CO₂ Spray Scrubbing with Monoethanolamine

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Abstract
CO₂ spray scrubbing was tested with Monoethanolamine (MEA) as an absorption solvent. The spray scrubber with a standard stripping unit attached was erected at a laboratory at the University of Leoben. The main goal of the design process was to create a pilot plant, which could operate under realistic conditions. This resulted in a maximum gas flow rate of 160 m³/h STP and a maximum liquid flow of 2 m³/h. The dimensions of the spray tower allow a gas velocity of 2.5 m/s. The concentration of the absorption liquid is adapted to common practice and set to 30w%. During the first test campaign with a single nozzle scrubber the functionality and the behavior of the test facility was studied. A continuous working pilot operation was successfully demonstrated. Some parameters like e.g. liquid to gas (L/G) ratio or different spray nozzles have been varied during these tests and their influence has been analyzed and evaluated. In addition, the influence of altered parameters on the energy required in the reboiler could be observed.

1. Introduction
Carbon Capture and Storage (CCS) is one of the most promising options to contribute to a stabilization of global greenhouse gas emissions. Among the three general CCS technologies (pre-combustion, post-combustion and oxy-fuel) post-combustion is the most advanced one, at the moment. In this process CO₂ is separated from the flue gas after the combustion step. The advantages of post combustion are the possibility of retrofitting existing power plants and the development status of the process, which is on the brink of marketability.

The absorption step is a crucial stage of the whole process. Hence, significant scientific effort has been made to improve absorption efficiency and to reduce operating and investment costs. At the moment most research focuses on the implementation of structured or random packed columns as absorber unit. Due to high gas flow rates of coal fired power plants ranging up to 4 million m³/h STP, the construction of such packed columns is technically challenging and still not fully solved. Up to now no packed columns have been built for high gas flow rates. Furthermore, a sufficient rate of CO₂-removal has not been demonstrated.

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yet in a single packed absorption column. Splitting the flue gas in two or more parallel streams to reduce the diameter of the columns, would lead to a significant increase in investment costs.

Another important aspect in column design is the total pressure drop in a column. High pressure drops, which are typical for packed columns, result in high energy consumption of the flue gas blower. Spray scrubbers on the other hand allow high flow rates and low pressure drops. Furthermore, spray scrubbing is a well-established industrial process for SO$_2$ removal. Absorption towers using spray scrubbing have already been built with diameters up to 24 meters and gas flow rates of around 4 million m$^3$/h STP. If these advantages can be transferred to the CO$_2$ sequestration process, spray scrubbing could be a potential solution to improve CCS technology.

As a consequence this project was developed in order to determine the potential of spray scrubbing for post combustion CO$_2$ capture. The scrubber used for the project has dimensions that allow tests under conditions close to reality. Previous studies focused mainly on hydraulic effects during absorption [1] or studied the use of caustic soda [2, 3]. This, however, is not suitable for large plants because of high material costs. The most promising absorption liquids at the moment are alkanolamines, especially Monoethanolamine (MEA). So far studies with MEA were only carried out under conditions not related to power plants [4].

Therefore, it was necessary to investigate CO$_2$ spray scrubbing with MEA as an absorption solvent. Spray scrubbing for CO$_2$ capture was tested in lab-scale at the University of Leoben. Some parameters were varied to find the achievable removal efficiency and to gain first experiences in CO$_2$ spray scrubbing. For the first test period the CO$_2$ scrubbing could only be tested with a single nozzle level and two different nozzle sizes. In future projects the CO$_2$ scrubbing will be tested with three nozzle levels, which will very likely increase the CO$_2$ absorption performance and with that the relevance and significance for industrial applications.

2. Spray Scrubbing test facility

The main advantage of the existing spray scrubber is its dimensions allowing fluid hydraulic conditions, which are very close to those in FGD spray scrubbers. This means that a maximum gas velocity of 2.5 m/s and a liquid to gas (L/G) ratio up to 34 L/m$^3$ could be reached. Due to the fast absorption kinetics and the large amount of available studies, 30w% MEA solution was chosen as the first absorption solvent. The existing SO$_2$ scrubber consists of the main components absorption column, sump tank, gas mixing station, blower, pumps and a gas analyzing station.

To allow continuous operation this plant had to be expanded with a desorption unit consisting of a desorption column and a reboiler mounted in the desorber sump tank. Furthermore, a central heat exchanger was necessary to reclaim energy from the recycled MEA solution. In addition a cooler unit was installed to regulate the temperature in the absorption column. Also various control and measuring devices had to be added. The whole process is illustrated as a schematic diagram in Figure 1.
Figure 1: Schematic diagram of the CO₂ spray scrubbing pilot plant

The gas mixing station creates a synthetic flue gas, in which the concentrations of N₂, O₂, SO₂, and CO₂ can be adjusted individually. A blower provides the desired gas flow rate of up to 160 m³/h. The flue gas enters the absorption column at the bottom and CO₂ is removed by the MEA solution which is injected at the column head with a maximum liquid flow rate of 2 m³/h. The loaded MEA solution leaves the absorber at the bottom and is collected in a separate sump. From there, it is pumped through the main heat exchanger to the top of the desorption column. In the heat exchanger energy is transferred from the lean to the rich MEA solution. In the desorption column the solution flows in counter current with the stripping steam produced in the reboiler through a random filling material of 25 mm Pall Rings. The regenerated lean MEA solution is pumped back through the main heat exchanger and finally adjusted to the absorption temperature in the cooler. In order to minimize operating costs, the bulk of the captured CO₂ is recycled to the gas mixing station. Therefore, the mixture of CO₂ and water vapor leaving the head of the desorption column is cooled in a plate type heat exchanger to condense the water. The two phases are then separated in a small vessel (Flash). The liquid water is returned to the reboiler tank to ensure a stable water balance and the gaseous CO₂ is returned to the gas mixing station. From this recycle stream a small partial flow is emitted to the environment in order to facilitate gas mixing. The whole process is logged and controlled using the National Instruments software “Lookout” and National Instruments modules are used to read signals from the sensors and to adjust valve settings and the electric heater unit. During the experiments it was necessary to check the concentration of the absorption liquid permanently and to recover MEA losses due to degradation via MEA make-up. Table 1 shows the process parameters of the used spray scrubber.

Table 1: Standard process parameter for spray scrubbing

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO₂ fraction in flue gas (dry)</td>
<td>14.0vol%</td>
</tr>
<tr>
<td>O₂ fraction in flue gas (dry)</td>
<td>6.0vol%</td>
</tr>
<tr>
<td>N₂ fraction in flue gas (dry)</td>
<td>80.0vol%</td>
</tr>
<tr>
<td>MEA concentration</td>
<td>30.0w%</td>
</tr>
<tr>
<td>max. gas flow rate</td>
<td>160 m³/h</td>
</tr>
<tr>
<td>max. liquid flow rate</td>
<td>2 m³/h</td>
</tr>
<tr>
<td>Temperature absorber</td>
<td>40°C</td>
</tr>
<tr>
<td>Temperature desorber</td>
<td>120°C</td>
</tr>
</tbody>
</table>
3. Results and Discussion

Two nozzle types (coarse and file) with different droplet sizes were investigated in this study. Furthermore, parameters like L/G ratio and superficial velocity were varied. The effect of a bubble column in combination with one spray nozzle was investigated too. Unfortunately the blower performance was too low to allow a bubble column higher than 20 cm due to the pressure drop. That’s why the positive effect of the bubble column could only be observed on a small scale. The following diagrams are showing the first results of the test facility. Figure 2 shows the dependency of the achieved CO₂ removal efficiency with the variation of the L/G ratio for different nozzles and the bubble column.

![Graph](image)

Figure 2: L/G ratio versus CO₂ removal efficiency; ▲ coarse spray nozzle (ds > 1000 µm); ● medium-fine spray nozzle (ds < 800 µm); ■ medium-fine spray nozzle with bubble column;

It is quite obvious that the bubble column supply increases the CO₂ removal efficiency only marginally. This is probably due to the low height of the bubble column. The higher pressure drop of the bubble column and the limiting power of the blower resulted in low gas flow rates leading to high L/G ratios. Furthermore, the coarse spray nozzle has little absorption rates (about 25%), but high L/G ratios (about 10.5 to 13 L/m³). Hence, only a few tests were performed with this nozzle.
Figure 3 depicts the correlation between the L/G ratio and the reboiler duty. The diagram shows that low L/G ratios lead to low reboiler duties of about 4 GJ/t CO₂. High gas flow rates and a high superficial velocity of about 2.5 m/s lead to little energy requirements for the regeneration of the absorption liquid. Higher L/G ratios, which were only possible due to low superficial velocities, lead to very high reboiler duties up to 14 GJ/t CO₂.

Figure 4 shows the dependency between the superficial velocity and the CO₂ removal efficiency. This illustrates that high superficial velocities lead to low CO₂ removal rates, which indicates a kinetic or a mass transfer problem in the spray scrubber. Probably, this problem could be solved with slower gas velocities or higher absorption columns with more than one spray nozzle to increase the residence time of the gas in the spray tower.
4. Outlook

Based on the results with the simplified single nozzle scrubber and the demonstration of a continuous working absorption/desorption cycle, the spray absorber will be modified and improved. The first test results show clearly that a suitable separation rate cannot be achieved with a single nozzle. Hence, three spray nozzles in series will be installed in the new and improved absorber. This will lead to a bigger active reaction surface and should enhance the mass transfer. Furthermore, a new blower will be installed to ensure higher gas flow rates and to overcome the bigger pressure drop. Therewith, a higher bubble column will also be tested at the bottom of the absorber to determine effects on the capture performance and the system behavior. A certain goal of this work is to determine the individual contributions of the different removal units and to identify whether it is possible to achieve the desired separation rates with different process modifications.

After achieving a stable and continuous mode of operation for the multiple spray scrubber and the active bubble layer, the apparatus could be split into a separate absorption and desorption unit to reduce the energy demand for stripping. Usually spray scrubbers need higher liquid to gas ratios compared to structured packed columns to provide a reasonable reaction surface. This high liquid flow rate reduces the differential liquid loading between the rich and the lean solvent and increases the energy consumption in the stripper, because the entire absorption solvent has to be heated in every cycle. Providing the necessary high liquid to gas ratio in the absorber, combined with a smaller split stream for the desorption unit, could possibly reduce the energy demand for the whole capture process. The highly regenerated split stream could be used in the top of the absorber to enhance the capture performance.

The main goal of the upcoming project is to vary different process parameters, including the L/G ratio, different spray nozzles, different heights of the active bubble layer, different split streams and to determine the best operation point of such an absorption unit. Furthermore various alterations can be made to the test procedure to: e.g. the CO$_2$ concentration in the flue gas can be raised up to 30vol% or SO$_2$ can be added as impurity.

5. Acknowledgment

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6. References